

A Comparison of Solar Wind and Estimated Solar System Xenon Abundances: A Test for Solid/Gas Fractionation in the Solar Nebula

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Significant fractionation of dust/gas from the original interstellar cloud during the formation of the solar system is a distinct possibility. Identification of such an effect would provide important clues to nebular processes. Fractionation of volatiles is not constrained by CI abundances and only for the most abundant ones by photospheric observations. The solar Xe elemental abundance is determined here via solar wind measurements from lunar ilmenites and normalized to Si by spacecraft data. The results are compared with estimated abundances assuming no fractionation, which are relatively well constrained for Xe by s-process calculations, odd-mass abundance interpolations, and odd-even abundance systematics. When corrected for solar wind/photospheric fractionation, the ^{136}Xe abundance given by surface layer oxidation of ilmenite from soil 71501, exposed within the last ~ 200 m.y., is 0.24 ± 0.09 normalized to $\text{Si} = 10^6$. This is indistinguishable from the estimates made assuming no solid/gas fractionation. A similar result was obtained for Kr by Wiens et al. (1991). Results from breccia 79035 ilmenite, exposed at least ~ 1 Gy ago, indicate that the solar wind Xe flux may have been significantly higher relative to other noble gases, perhaps due to more efficient Xe ionization. If this is true, fluxes of C and S, which have similar first ionization potentials to Xe, should also be higher in the ancient solar wind from the same time period, though such variations have not been observed.

FRACTIONATION IN THE SOLAR NEBULA

A detailed knowledge of the average elemental abundances in the solar system is crucial to our understanding of its origin and evolution. Specifically, depletions of certain types of elements relative to an expected original abundance in the parent interstellar cloud from which the solar nebula formed would give clues to whether various possible fractionation processes were active in the gravitational collapse leading to the formation of the Sun and planets. Even limits on the extent of these fractionations will help us understand the processes. Fractionation could occur in a variety of ways: by electromagnetic processes, albedo variations (on dust grains), different atomic mass or density, or with differences in volatility. In this paper we compare estimates of the solar system Xe/Si ratio, obtained independently of any solar data, to that determined from the solar wind by a combination of lunar and spacecraft data. These independent determinations could differ if there are fractionation processes, e.g., solid/gas, in the formation of the Sun from the original parent molecular cloud.

The Sun's formation can be conceptualized as a two-stage process, with the first stage consisting of formation of a central condensation out of the parent giant molecular cloud, and the second stage consisting of passage through a T Tauri phase and onto the main sequence. The CI chondrite abundance pattern, representing a point in time after stage 1, defines a smooth

abundance curve for heavy ($A > 60$) odd-mass nuclei (Anders and Grevesse, 1989; Burnett et al., 1989), a regularity that must be nucleosynthetic in origin (Suess, 1947). The CI abundance pattern thus applies to stage 1, assuming that all significant nucleosynthesis occurred prior to that time. Further, all elements included in these systematics must have been essentially quantitatively ($\geq 70\%$) condensed at the formation location of the CI chondrites. Specifically, this includes all elements from Ni to Pb except Kr, Xe, and possibly Br (Burnett et al., 1989). The present solar composition is given by photospheric abundances, which agree with CI chondrite abundances within 10-30% for many elements (e.g., Anders and Grevesse, 1989; Grevesse, 1991). The original starting materials, that is, the relative abundances of the parent molecular cloud, are accurately modeled in the high mass range ($A = 100-200$) by the classical s-process theory (e.g., Käppeler et al., 1989). There is general agreement among these three sources of nonvolatile element abundances, so that among the solid elements very little fractionation (probably $< 30\%$) could have taken place during the formation of the Sun. [There is no unique distinction between volatile (gas) and nonvolatile (solid) elements. The partitioning of a given element between gas and condensed phases obviously depends on physical conditions, primarily temperature. Our discussion is relevant to environments in which Kr and Xe are in the gas phase, with less-volatile elements generally in the solid phase.]

SOLID/GAS FRACTIONATION IN THE SOLAR NEBULA

The story for volatile elements is quite different. Volatiles have been depleted in the CI chondrites and, for the most part, are not observable in the solar photosphere due to the high first excited atomic state of gases, resulting in the absence of characteristic absorption lines. Therefore, a relatively large solid/gas fractionation in the Sun would not be obvious by any means studied so far.

Some of the mechanisms that would affect solid/gas ratios, following *Wiens et al.* (1991), include:

1. Fractionation due to intense stellar winds early in solar history. Efficient sinks for angular momentum and magnetic fields are needed during collapse of the solar nebula, and substantial mass loss appears to occur during and immediately preceding a T Tauri phase (e.g., *Lada and Shu*, 1990). By analogy with the present-day solar wind, a fractionation by first ionization potential (FIP) might result because of the preferential ionization of elements with low FIP. This might be accomplished even with a small degree of ionization, given that the mass loss would occur over relatively large distances and timescales. Since volatile elements have relatively high FIPs this mechanism effectively leads to low nonvolatile/volatile element ratios in the Sun even though volatility per se was not the important parameter. The FIP fractionation factor in the solar wind today is ~ 4 , and far higher factors have been suggested (*Ott and Begemann*, 1990; *Lewis et al.*, 1990) to explain elemental enrichments in interstellar dust grains. While the present solar wind flux extrapolated over the age of the solar system should have depleted the remaining nonvolatile elements by less than 1% relative to the initial volatile/nonvolatile inventory in the solar convective zone, volatile/nonvolatile fractionation could be significant if the Sun lost a substantial fraction of its original mass during an early T Tauri stage. However, this fractionation only affects the gas phase, so that higher temperatures (>2000 K) would be required for Xe/Si to be fractionated in this way.

2. A variation of (1) at lower temperature with Si in grains but Xe and Kr in the gas phase. A low level of ionization and separation of the bulk material from originally imbedded magnetic fields would produce a fractionation of gas ions (including Kr and Xe) from solids. This mechanism may have been active in the interstellar cloud long before ignition of the Sun, since magnetohydrodynamic processes are thought to play a crucial role in causing local inhomogeneities leading to stellar formation (*Lada and Shu*, 1990).

3. Solid/gas fractionation due to the Poynting-Robertson effect, which causes solid particles to slowly spiral toward the Sun. Today only solids spiral toward the Sun. The mass flux is insignificant relative to the mass of the solar convection zone, and it is not clear whether the atoms in the incoming particles are accreted to the Sun or vaporized and swept out with the solar wind. However, this mechanism could have been significant in the early solar system when unaccreted materials were abundant, gas drag possibly significant, and the solar surface temperature much lower. In addition to electromagnetic drag, a strong early solar wind may have caused an enhanced corpuscular radiation analogue of the Poynting-Robertson effect on grains smaller than $\sim 1 \mu\text{m}$ (*Burns et al.*, 1979).

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5. Similar to (4) but with fractionation in the galaxy as a whole due to removal of all elements condensable at temperatures of ~ 20 -50 K by large-scale comet and ice formation. As proposed by *Tinsley and Cameron* (1974), only H and He would not be condensed.

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UNFRACTIONATED (ORIGINAL) XENON ABUNDANCE

For xenon, interpolation between the surrounding odd-mass isotopes is not as well constrained as Kr because Te and Xe lie on an r-process peak rather than a slowly decreasing part of the abundance curve. Fortunately, Xe has two isotopes that are shielded from the r-process contributions and produced almost exclusively by the s-process. Contemporary s-process theory provides an excellent quantitative fit to the observed CI chondrite abundances for nonvolatile s-only nuclei in the $A > 100$ mass range, the so-called "main component" of the classical s-process theory of *Käppeler et al.* (1989). The theory predicts that s-process abundances are described by a smooth variation of $N\sigma$, where N is the abundance of the given isotope and σ is the laboratory-derived neutron absorption cross section at the relevant energy. S-process abundances are thus $(N\sigma)_{\text{theory}}/\sigma$. If one assumes that neutron capture cross sections at $kT = 25$ -30 keV are applicable, there are only two adjustable parameters (a constant and one exponential parameter in the neutron fluence distribution incident on the seed ^{56}Fe) in the *Käppeler et al.* (1989) calculations of the main s-process. The s-process results are compared with nonvolatile CI abundances for s-only isotopes from 96-204 amu in Fig. 1. In almost every case the agreement is within 20-30%, enabling comparison on a linear scale. There is a minor complication in that estimated abundances of ^{130}Xe and ^{128}Xe were used in the overall fit by *Käppeler et al.* (1989). However, as these are only two out of the whole dataset being fitted, it is safe to assume that the predicted $(N\sigma)$ would be the same if the two s-only Xe isotopes were excluded from the fit. We there-

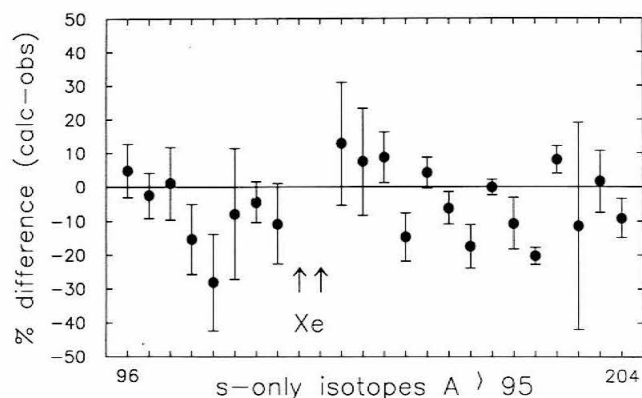


Fig. 1. Comparison of s-process abundances calculated from ($N\sigma$) and σ values tabulated by (Käppeler et al., 1989) with CI chondrite abundances from Anders and Grevesse (1989) for s-only isotopes of nonvolatile elements in the high-mass range. The agreement is within 10–30% in most cases, justifying the use of s-process Xe abundance calculations as representative of the original unfractionated Xe abundance.

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(Wieler et al., 1986) or low-temperature oxidation (Frick et al., 1988). Only the latter method has produced data for heavy noble gases (Kr and Xe). We use data from the only two lunar ilmenite separates subjected to this procedure to date, 71501 (Frick et al., 1988) and 79035 (Becker and Pepin, 1989). Their light noble gases are compared in Fig. 2 with the solar wind.

Nitrogen/Noble Gas Ratios

Although the light noble gas patterns are nearly identical to the solar wind in the 71501 and 79035 ilmenite surface layers oxidized at low temperatures (Fig. 2), the relative nitrogen abundances, both of the near-surface and the sample as a whole, are higher than expected in the solar wind by about an order of magnitude (Frick et al., 1988; Becker and Pepin, 1989). This presents a possible objection to considering surface-sited noble gases as representative of the solar wind. It appears that either noble gases are inefficiently implanted or there is an extraneous source of nitrogen implanted at

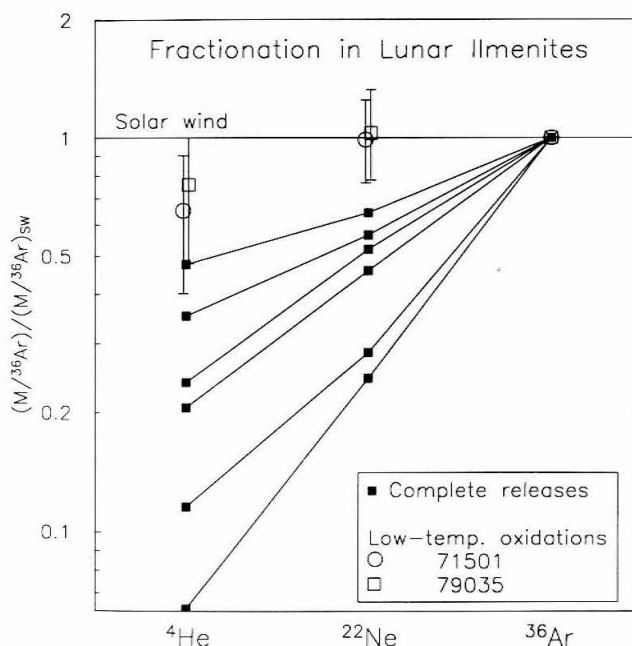


Fig. 2. Helium and Ne abundances from lunar ilmenite separates normalized to the solar wind and to Ar. Filled squares represent total gas amounts from six different samples measured by Hintenberger et al. (1974), Frick et al. (1988), and Becker and Pepin (1989), which are fractionated relative to the solar wind. Open symbols represent gases released from only the surface layers by low-temperature oxidation of breccia 79035 (Becker and Pepin, 1989) and the more recently exposed soil 71501 (Frick et al., 1988). Notice that the $^{22}\text{Ne}/^{36}\text{Ar}$ ratio is unfractionated in both samples, and it is assumed that the heavier noble gases are also unfractionated. Normalization to the solar wind provides a better fit, particularly for $^{22}\text{Ne}/^{36}\text{Ar}$, than normalization to estimated solar abundances used in previous comparisons (Frick et al., 1988; Becker and Pepin, 1989). The major source of uncertainties is in the long-term average solar wind ratios (Bochsler, 1987).

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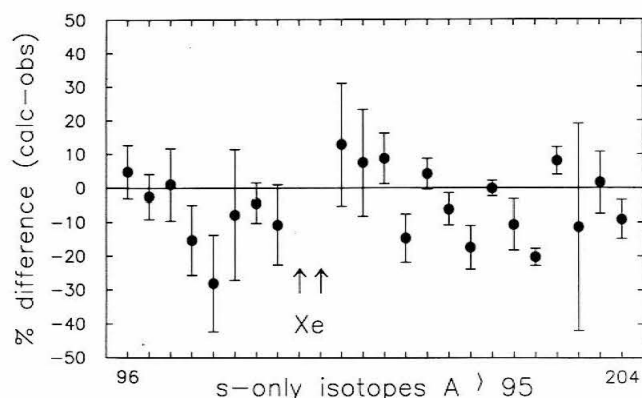


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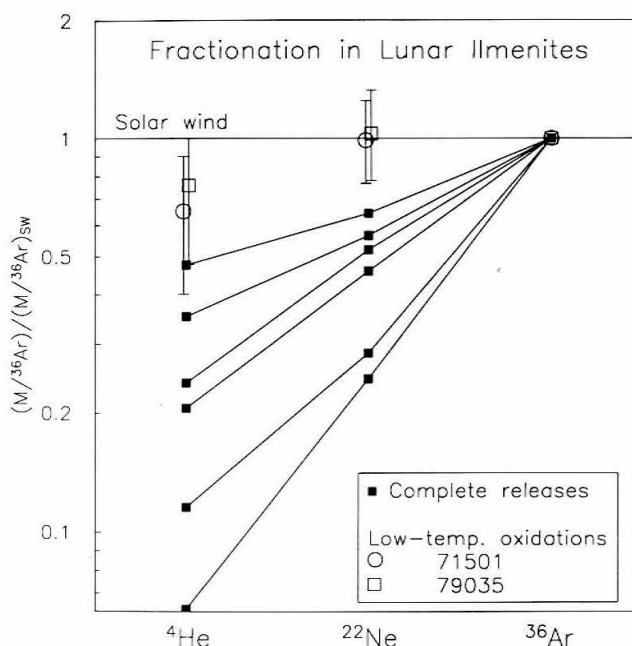


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energies of similar order of magnitude to the solar wind. Both of these options have been argued against (*Becker and Pepin, 1989*). A possible third alternative (*Frick et al., 1988*) is that nitrogen diffuses less readily than the noble gases, so that N from many exposure episodes tends to remain in the surface reservoir, while noble gases do not. A firm resolution to this problem is needed to completely validate the results presented here.

Antiquities

Judging in each case from the $^{40}\text{Ar}/^{36}\text{Ar}$ ratios (e.g., *Kerridge, 1980; Wieler et al., 1983*), the solar wind exposure of surface soil 71501 probably occurred within the last several hundred million years, while soil breccia 79035 was most likely exposed at least ~ 1 Gy before present. The low $\delta^{15}\text{N}$ values measured in 79035 (*Clayton and Thiemens, 1980; Becker and Pepin, 1989*) suggested that it might be significantly older, up to 2-2.5 Gy (*Clayton and Thiemens, 1980*) by the assumption that $\delta^{15}\text{N}$ decreased with age before present. However, based on spallation product considerations, a younger age (between 960 and 1240 m.y.) has recently been suggested by *Benkert et al. (1991)*. This must be a lower limit to 79035 that, if true, would imply that the lowest $\delta^{15}\text{N}$ and highest relative Xe abundance (discussed below) do not belong to the oldest samples (i.e., samples with the highest $^{40}\text{Ar}/^{36}\text{Ar}$), but perhaps represent an anomalous spike in solar wind composition.

SOLAR WIND XENON

Low-temperature ilmenite oxidations give ratios of ^{130}Xe relative to ^{20}Ne and ^{36}Ar . These are normalized to Si using spacecraft detector ratios of $\text{Ne}/\text{O} = 0.17 \pm 0.02$ (*Bochsler and Geiss, 1989; Bochsler et al., 1986*) and $\text{Si}/\text{O} = 0.19 \pm 0.04$ (*Bochsler, 1989*), and, where ilmenite ratios relative to ^{36}Ar are used, the Apollo solar wind foil ratio $^{36}\text{Ar}/\text{Ne} = 0.0205 \pm 0.0050$ (*Geiss et al., 1972; Bochsler and Geiss, 1977*). The data and results are shown in Table 1 for Xe in ilmenite from 71501 and 79035. The solar wind Xe abundances calculated by this method are well below solar system estimates given above. A

similar result was found by *Wiens et al. (1991)* for Kr. The simplest explanation for this difference is that the solar wind Xe/Si ratio is fractionated relative to the photosphere because of the difference in FIP. The first ionization potential of Xe is 12.1 V; the Si FIP is 8.1 V. All observed elements with FIPs greater than ~ 10 V are depleted in the solar wind and solar energetic particles (SEPs) by an approximately constant value relative to lower FIP elements. This is illustrated schematically by the solid line in Fig. 3, after the model of *Steiger and Geiss*

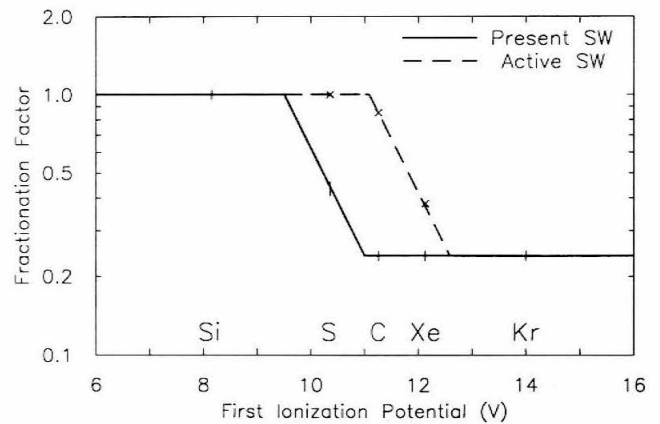


Fig. 3. Schematic fractionation patterns for selected elements in the solar wind relative to the photosphere and normalized to Si. The solid line with tic marks for the relevant elements is after the model of *Steiger and Geiss (1988)* for the present-day solar wind. Most of the nonvolatile elements plot on the higher portion of the line near Si, while most of the volatile elements lie near Kr. The dashed line with x's represents a proposed mechanism to explain why Xe released by low-temperature oxidation of 79035 ilmenite (*Becker and Pepin, 1989*) is a factor of 1.6 higher relative to the other noble gases than the recent solar wind, as measured in 71501 ilmenite (*Frick et al., 1988*). It is proposed that the FIP cutoff in the ancient solar wind was higher than at present. This simplistic scenario, discussed in the final section of the paper, predicts a factor of nearly 4 higher relative solar wind C flux for the active period.

TABLE 1. Data used for calculation of the solar wind ^{130}Xe abundances.

Sample	Ilmenites		Directly-measured SW		^{130}Xe (Si= 10^6)
	$^{130}\text{Xe}/^{20}\text{Ne}$	$^{130}\text{Xe}/^{36}\text{Ar}$	$^{20}\text{Ne}/\text{Si}$	$^{36}\text{Ar}/\text{Si}$	
71501	6.82×10^{-8} ± 0.31		0.83 ± 0.20		0.057 ± 0.014
71501		3.11×10^{-6} ± 0.21		0.0183 ± 0.0063	0.057 ± 0.020
79035	1.09×10^{-7} ± 0.13		0.83 ± 0.20		0.090 ± 0.024
79035		5.05×10^{-6} ± 0.45		0.0183 ± 0.0063	0.092 ± 0.033

Lunar ilmenite data are from *Frick et al. (1988)* and *Becker and Pepin (1989)*; solar-wind $^{20}\text{Ne}/\text{Si}$ and $^{36}\text{Ar}/\text{Si}$ are from *Geiss et al. (1972)*, *Bochsler and Geiss (1977)*, *Bochsler (1989)*, *Bochsler et al. (1986)*, and *Bochsler and Geiss (1989)*. Two calculations are given for each sample, one using the ilmenite $^{130}\text{Xe}/^{20}\text{Ne}$ ratio and the other the $^{130}\text{Xe}/^{36}\text{Ar}$ ratio.

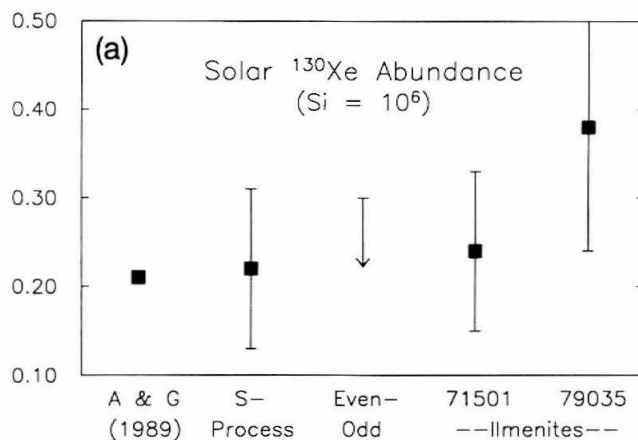
(1989). The FIP fractionation factor is still poorly constrained. Solar wind fractionation is much more uncertain than SEP fractionation, for which the most recent estimate is 4.4 (Grevesse, 1991); fractionation of the two types of particles need not be identical. The solar wind depletion of Xe relative to Si should be well within a conservative estimate of 4.2 ± 1.5 (Steiger and Geiss, 1989; Anders and Grevesse, 1989) used by Wiens et al. (1991) for Kr.

The solar wind ^{130}Xe results are given in Table 2 after averaging the two methods for each sample and correcting for fractionation between the photosphere and the solar wind by the above factor of 4.2. The results are shown in Fig. 4a alongside independent estimates of the solar system Xe abundance that assume no fractionation. The estimates are given by (1) interpolation of both even and odd isotopes with CI abundances of Te, I, Cs, and Ba by Anders and Grevesse (1989), (2) classical main-component s-process calculations (Käppeler et al., 1989), and (3) the even-odd constraint

TABLE 2. Solar xenon abundances.

Source	^{130}Xe
71501 ilmenite	0.24 ± 0.09
79035 ilmenite	0.38 ± 0.14
Anders and Grevesse (1989)	0.21
S-process (Käppeler et al., 1989)	0.22 ± 0.09
Even-odd abundances	≤ 0.30

The first two lines give the ^{130}Xe data from Table 1, averaged for their respective samples and corrected for FIP fractionation by a factor of 4.2 ± 1.5 . For comparison, ^{130}Xe estimates from CI interpolations, s-process systematics, and even-odd abundance considerations are given relative to $\text{Si} = 10^6$. The bottom three estimates assume no solid/gas fractionation.



described above. The more recently-exposed ilmenite, 71501, is nearly identical to the estimates, consistent with no, or at least very little, solid/gas fractionation. Uncertainty for the 79035 datum overlaps the 71501 uncertainty, but the nominal value is above even the even-odd mass constraint in Fig. 4a. Most of the uncertainty is from the Si normalization and the FIP fractionation factor. Relative to 71501, the high Xe abundance in the 79035 ilmenite surface layer is highly significant (Becker and Pepin, 1989). The 79035 data point in Fig. 4a could thus be construed as evidence for enrichment of gas relative to solids in the Sun. However, the ^{83}Kr results for both samples (Wiens et al., 1991), shown in Fig. 4b after similar correction for FIP fractionation, agree with the CI-interpolated abundances, showing no evidence for enrichment of gases in the Sun.

Since Xe from 71501 and Kr from both samples are very near the unfractionated abundance estimates, our conclusion is that, within the uncertainties, the Sun's solid/gas ratio is unfractionated from the original presolar material. This conclusion is only accurate to a factor of 2 at present. It is important to test for solid/gas fractionation at higher levels of accuracy, based on improved observations of solar wind abundances.

The possibility of chemical fractionation of solar matter relative to the parent molecular cloud deserves more attention than it has been hitherto given. Of the works cited earlier, the calculations of Joss (1974) focused on one specific mechanism: comet accretion to the Sun. Joss (1974) concluded that such accretion could be important, but his idea has been largely ignored. Tinsley and Cameron (1974) considered an even grander-scale application of solid/gas fractionation: to the galaxy as a whole and the issue of low galactic heavy element

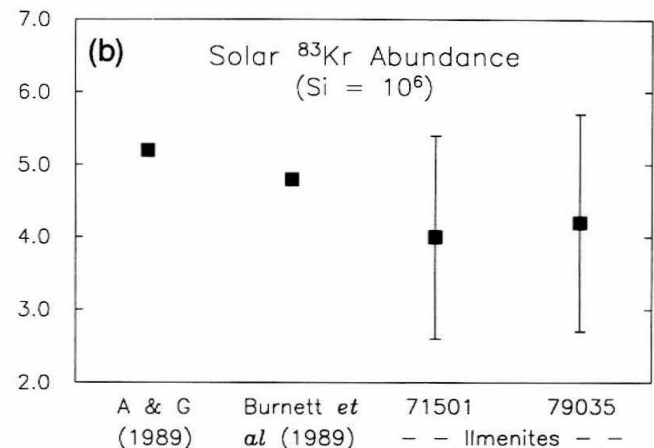


Fig. 4. (a) Solar ^{130}Xe abundances determined from solar wind trapped in ilmenites 71501 and 79035, and estimates from Table 2 given by interpolation (Anders and Grevesse, 1989), s-process calculations (Käppeler et al., 1989), and the even-odd constraint, all three of which assume no solid/gas fractionation. Solar wind data from the 71501 ilmenite, which are sensitive to solid/gas fractionation, are in agreement. However, the more ancient solar wind captured in 79035 is nominally high. Most of the uncertainty is from the solar wind Si normalization and from the solar wind fractionation factor. The differences in Xe/Ar and Xe/Ne released from the two ilmenites are significant, and suggest that a modification has occurred in solar wind Xe over time (Becker and Pepin, 1989). (b) Solar ^{83}Kr estimates assuming no solid/gas fractionation, and solar abundances derived similarly to the Xe. These are unchanged from Wiens et al. (1991) and are repeated here for completeness. The solar-wind-derived Kr data are within uncertainty of estimates. A value of $^{83}\text{Kr} = 5.2$ ($\text{Si} = 10^6$) was estimated by Anders and Grevesse (1989); the interpolated value from Burnett et al. (1989) is 4.75.

abundances. Their suggestion has not found favor among galactic astronomers. As a solar system process, it deserves serious consideration, although in the literal version proposed by the authors, only H and He would be left in the gas phase. Because CI interpolations are not possible for H and He, such fractionation would be very difficult to establish, compared to the more favorable cases for Kr and Xe that we have discussed. Fractionation by FIP, which is effectively solid/gas fractionation, has not to our knowledge been discussed in the context of the solar system, even though it may be the most likely fractionation mechanism. Ironically, it may be that the lack of fractionation, assuming that this could be demonstrated at a higher level of accuracy than at present, is the most powerful constraint on physical processes active during the evolution of the Sun from the parent molecular cloud. Such lack of fractionation would then be a required prediction of specific theories.

Xenon Abundance Modifications in 79035

Because of its magnitude (a factor of 1.6 greater than 71501 with no hint of concomitant Kr, Ar, or Ne increases) and because the higher relative Xe abundance persists in the high-temperature fraction (Becker and Pepin, 1989), it is unlikely that the higher Xe abundance in 79035 is simply an artifact of a previous regolith exposure cycle from which the Xe diffused more slowly than the other gases. However, this remains a possibility, as it does for the nitrogen.

An alternative view is that the Xe in 79035 experienced the secular solar wind flux enhancement proposed for other samples with great antiquities (e.g., Kerridge, 1980). If this is the case, the even-odd abundance constraint indicates that either all elements in the Xe mass region were enhanced or, more likely, the solar wind/photosphere FIP fractionation factor was modified. The Xe FIP, at 12.1 V, is the lowest of the stable noble gases, and is thus the most likely to be altered during a period of unusually high solar activity.

Details on how this could have occurred are poorly known. It is possible that, during such a period, the relative contribution of transient solar wind associated with coronal mass ejections was higher than the present ~5% relative to the steady-state solar wind. The relative abundance of He, which has the highest FIP, rises to 20% or greater in the transient solar wind (Neugebauer, 1981). However, some elements with low FIP, such as Fe, are also enhanced, and the levels of less-abundant elements are still not known, so it is not clear what overall effect a more significant transient solar wind contribution would have on long-term average abundances.

Another possible explanation may be that a modification of the parameters in the photosphere led to a higher cutoff as a function of FIP for the fractionation of the steady-state solar wind, as illustrated by the dashed line in Fig. 3. This would explain why Kr, with a higher FIP than Xe, does not show an enhancement in 79035. Perhaps a model similar to that developed by Steiger and Geiss (1989), which fits the change in ionization efficiency at ~10 V, could be made to fit such a change at ~12 V instead, as shown in the figure. More realistically, the early active Sun may have had a wide range of FIP cutoffs leading to systematic fractionations among medium- to high-FIP elements.

A change in solar activity strong enough to alter the relative Xe abundance should also affect several other elements with similar FIPs. The most abundant of these are S and C, which have FIPs of 10.4 and 11.3 V respectively. Carbon is presently fractionated by approximately the same factor as volatile elements with higher FIPs (Steiger and Geiss, 1989), as shown by the tic mark on the solid line in Fig. 3. If Xe were enhanced by a factor of 1.6 during exposure of 79035, C should be enhanced by at least that much, and could be up to a factor of ~4 higher, as shown along the dashed line (Fig. 3), if C was nearly unfractionated relative to Si. Sulfur, which is presently fractionated by an intermediate value (Steiger and Geiss, 1989), would be enriched by a correspondingly smaller amount. Data on solar wind S and unambiguous concentrations of solar wind C in lunar material are not available. As with the noble gases, bulk concentrations almost certainly will not give true solar wind abundances. It would be necessary to measure the most recent surface loadings of solar wind C and S. However, for both elements the situation is not as simple as for noble gases. Terrestrial surface contamination and volatilized C and S from micrometeorite bombardment make surface layer solar wind analysis very difficult at best.

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REFERENCES

- Anders E. and Grevesse N. (1989) Abundances of the elements: Meteoritic and solar. *Geochim. Cosmochim. Acta*, 53, 197-214.
- Becker R. H. and Pepin R. O. (1989) Long-term changes in solar wind elemental and isotopic ratios: A comparison of two lunar ilmenites of different antiquities. *Geochim. Cosmochim. Acta*, 53, 1135-1146.
- Benkert J. P., Kerridge J. F., Kim J. S., Kim Y., Marti K., Signer P., and Wieler R. (1991) Evolution of isotopic signatures in lunar-regolith nitrogen: Noble gases and N in ilmenite grain-size fractions from regolith breccia 79035 (abstract). In *Lunar and Planetary Science XXII*, pp. 85-86. Lunar and Planetary Institute, Houston.
- Bochsler P. (1989) Velocity and abundance of silicon ions in the solar wind. *J. Geophys. Res.*, 94, 2365-2373.
- Bochsler P. and Geiss J. (1977) Elemental abundances in the solar wind. *Trans. Intl. Astron. Union, XVII, Proc. 16th Gen. Assembly* (E. A. Miller and A. Jappel, eds.), pp. 120-123.
- Bochsler P. and Geiss J. (1989) Composition of the solar wind. In *Solar System Plasma Phys., Geophys. Mon.*, 54 (J. H. Waite Jr., J. L. Burch, and R. L. Moore, eds.), pp. 133-141.
- Bochsler P., Geiss J., and Kunz S. (1986) Abundances of carbon, oxygen, and neon in the solar wind during the period from August 1978 to June 1982. *Solar Phys.*, 103, 177-201.
- Burnett D. S., Woolum D. S., Benjamin T. M., Rogers P. S. Z., Duffy C. J., and Maggione C. (1989) A test of the smoothness of the elemental abundances of carbonaceous chondrites. *Geochim. Cosmochim. Acta*, 53, 471-481.
- Burns J. A., Lamy P. L., and Soter S. (1979) Radiation forces on small particles in the solar system. *Icarus*, 40, 1-48.
- Clayton R. N. and Thiemens M. H. (1980) Lunar nitrogen: Evidence for secular change in the solar wind. In *The Ancient Sun: Fossil Record in the Earth, Moon and Meteorites* (R. O. Pepin, J. E. Eddy, R. B. Merrill, eds.), pp. 463-473. Pergamon, New York.

- Frick U., Becker R. H., and Pepin R. O. (1988) Solar wind record in the lunar regolith: Nitrogen and noble gases. *Proc. Lunar Planet. Sci. Conf. 18th*, pp. 87-120.
- Geiss J., Bühler F., Cerutti H., Eberhardt P., and Filleux C. (1972) Solar wind composition experiment. In *Apollo 16 Preliminary Science Report*, pp. 14-1 to 14-10. NASA SP-315.
- Grevesse N. (1991) Solar abundances: The reference system. In *Evolution of Stars: The Photospheric Abundance Connection* (G. Michaud and A. Tutukov, eds.), pp. 63-69. Kluwer, Dordrecht.
- Hintenberger H., Weber H. W., and Schultz L. (1974) Solar, spallogenic, and radiogenic rare gases in Apollo 17 soils and breccias. *Proc. Lunar Sci. Conf. 5th*, pp. 2005-2022.
- Joss P. C. (1974) Are stellar surface heavy-element abundances systematically enhanced? *Astrophys. J.*, *191*, 771-774.
- Käppeler F., Beer H., and Wisshak K. (1989) S-process nucleosynthesis—nuclear physics and the classical model. *Rep. Prog. Phys.*, *52*, 945-1013.
- Kerridge J. F. (1980) Secular variations in composition of the solar wind: Evidence and causes. In *The Ancient Sun: Fossil Record in the Earth, Moon and Meteorites* (R. O. Pepin, J. E. Eddy, and R. B. Merrill, eds.), pp. 475-489. Pergamon, New York.
- Lada C. J. and Shu J. H. (1990) The formation of sunlike stars. *Science*, *248*, 564-572.
- Lewis R. S., Amari S., and Anders E. (1990) Meteoritic silicon carbide: Pristine material from carbon stars. *Nature*, *348*, 293.
- Neugebauer M. (1981) Observations of solar wind helium. *Fund. Cosmic Phys.*, *7*, 131-199.
- Ott U. and Begemann F. (1990) Discovery of s-process barium in the Murchison meteorite. *Astrophys. J.*, *353*, L57-L60.
- Steiger R. and Geiss J. (1989) Supply of fractionated gases to the corona. *Astron. Astrophys.*, *225*, 222-238.
- Suess H. (1947) Über kosmische Kernhäufigkeiten I. Mitteilung: Einige Häufigkeitsregeln und ihre Anwendung bei der Abschätzung der Häufigkeitswerte für die mittelschweren und schweren Elemente. II. Mitteilung: Einzelheiten in der Häufigkeitsverteilung der mittelschweren und schweren Kerne. *Z. Naturforsch.*, *2a*, 311-321, 604-608.
- Tinsley B. M. and Cameron A. G. W. (1974) Possible influence of comets on the chemical evolution of the galaxy. *Astrophys. Space Sci.*, *31*, 31-35.
- Wieler R., Etique P., Signer P., and Poupeau G. (1983) Decrease of the solar flare/solar wind flux ratio in the past several aeons deduced from solar neon tracks in lunar soil plagioclases. *Proc. Lunar Planet. Sci. Conf. 13th*, in *J. Geophys. Res.*, *88*, A713-A724.
- Wieler R., Baur H., and Signer P. (1986) Noble gases from solar energetic particles revealed by closed system stepwise etching of lunar soil minerals. *Geochim. Cosmochim. Acta*, *50*, 1997-2017.
- Wiens R. C., Burnett D. S., Neugebauer M., and Pepin R. O. (1991) Solar-wind krypton and solid/gas fractionation in the early solar nebula. *Geophys. Res. Lett.*, *18*, 207-210.